

**CO<sub>2</sub> emissions trends in the Los Angeles megacity**

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# Toward consistency between bottom-up CO<sub>2</sub> emissions trends and top-down atmospheric measurements in the Los Angeles megacity

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## Abstract

Large urban emissions of greenhouse gases result in large atmospheric enhancements relative to background that are easily measured. Using CO<sub>2</sub> mole fractions and  $\Delta^{14}\text{C}$  and  $\delta^{13}\text{C}$  values of CO<sub>2</sub> in the Los Angeles megacity observed in inland Pasadena (2006–2013) and coastal Palos Verdes peninsula (autumn 2009–2013), we have determined time series for CO<sub>2</sub> contributions from fossil fuel combustion for both sites and broken those down into contributions from petroleum/gasoline and natural gas burning for Pasadena. We find a 10% reduction in Pasadena CO<sub>2</sub> emissions from fossil fuel combustion during the Great Recession of 2008–2010, which is consistent with the bottom-up inventory determined by the California Air Resources Board. The isotopic variations and total atmospheric CO<sub>2</sub> from our observations are used to infer seasonality of natural gas and petroleum combustion. For natural gas, inferred emissions are out of phase with the seasonal cycle of total natural gas combustion seasonal patterns in bottom-up inventories but are consistent with the seasonality of natural gas usage by the area's electricity generating power plants. For petroleum, the inferred seasonality of CO<sub>2</sub> emissions from burning petroleum is delayed by several months relative to usage indicated by statewide gasoline taxes. Using the high-resolution Hestia-LA data product to compare emissions from parts of the basin sampled by winds at different times of year, we find that variations in observed fossil fuel CO<sub>2</sub> reflect seasonal variations in wind direction. The seasonality of the local CO<sub>2</sub> excess from fossil fuel combustion along the coast, on Palos Verdes peninsula, is higher in fall and winter than spring and summer, almost completely out of phase with that from Pasadena, also because of the annual variations of winds in the region. Variations in fossil fuel CO<sub>2</sub> signals are consistent with sampling the bottom-up Hestia-LA fossil CO<sub>2</sub> emissions product for sub-city source regions in the LA megacity domain when wind directions are considered.

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# 1 Introduction

Carbon dioxide is the most important greenhouse gas (GHG) contributing to current global warming, contributing 64 % of the total radiative forcing, according to the IPCC AR5 report (IPCC, 2013) and comprising 82 % of GHG emissions (NRC, 2010). The global average mole fraction of CO<sub>2</sub> has increased approximately 40 % since pre-industrial times due to anthropogenic emissions (IPCC, 2013). Since the proportion of the world's emissions from megacities (urban regions with more than 10 million inhabitants) is out of proportion with their small surface area (EDGAR, 2009; IEA, 2008), quantifying these emissions is essential if we are to work aggressively toward their reduction (Duren and Miller, 2012). As a consequence of global warming mitigation, reducing CO<sub>2</sub> emissions could reduce air pollution mortality, which is correlated with increased CO<sub>2</sub> levels (Jacobson, 2008). Identifying the sources of emissions is a major first step in understanding and mitigating anthropogenic contributions. In cities, these CO<sub>2</sub> sources often dominate over the normally predominant natural source of the biosphere, at least during certain seasons (e.g., Pataki et al., 2003; Widory and Javoy, 2003; Newman et al., 2013, 2008; Lopez et al., 2013; Turnbull et al., 2011, 2015; Vardag et al., 2015). The most common method of inventorying CO<sub>2</sub> emissions from human activities is through bottom-up reporting by governmental agencies, following IPCC methods (IPCC, 2013). This requires self-reporting and may not be reliable in some parts of the world. A more recent, scientifically-based bottom-up approach has been pioneered through the Vulcan and Hestia projects (Gurney et al., 2009, 2012). These efforts combine multiple streams of data such as air pollution reporting, demographics, property tax data, and traffic monitoring, to arrive at what is proving to be a much more accurate and space/time detailed estimate of fossil fuel CO<sub>2</sub> emissions. The Vulcan Project accomplished fossil fuel CO<sub>2</sub> emission estimation for the whole US at spatial scales of 10 km every hour of the year 2002, with updated years expected by the end of 2015. Hestia is specifically focused on the urban domain and has accomplished estimation down to the individual building and street segment scale for four

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cities (Indianapolis, IN; Salt Lake City, UT; Los Angeles basin, CA; Phoenix, AZ) with work ongoing in Baltimore, MD (Gurney et al., 2012, Patarasuk et al., in prep.; Rao et al., 2015). Both of these detailed data products are available for selected cities in the United States, facilitating top-down emissions quantification through long-term ambient air monitoring (Duren and Miller, 2012; Gurney et al., 2015). Trends in CO<sub>2</sub> emissions must be monitored precisely in order to evaluate progress towards mandated emission reductions. As an example, the California Global Warming Solutions Act of 2006 (Assembly Bill 32) requires reduction of greenhouse gas emissions to 1990 levels by 2020, a reduction of about 15 %. Indeed, now is the time to document the current level of emissions, as governments begin to implement strategies to reduce emissions (e.g., California's Cap-and-Trade Program and Low Carbon Fuel Standards) and want to be able to assess their efficacy.

Within megacities, atmospheric CO<sub>2</sub> concentrations are often highly elevated relative to the regional background due to locally emitted carbon dioxide. This excess can be analyzed for its isotopic composition to help attribute the local emissions to specific processes. Radiocarbon (<sup>14</sup>C) analyses give quantitative information as to the proportions of CO<sub>2</sub> resulting from combustion of ancient sources of carbon (fossil fuels) relative to sources incorporating modern carbon, such as the biosphere (e.g., Levin et al., 2003; Levin and Roedenbeck, 2008; Turnbull et al., 2009), because of its short half-life of 5730 years. The stable isotopes of carbon can be used to separate sources with differing values, such as natural gas and petroleum combustion, with the <sup>13</sup>C/<sup>12</sup>C ratio of natural gas typically being lower than that of petroleum (e.g., Keeling, 1961, 1958; Newman et al., 2008, 2013; Pataki et al., 2003; Widory and Javoy, 2003; Djuricin et al., 2010; Moore and Jacobson, 2015), although there can be overlap between petroleum combustion and biological respiration. Therefore, if we know the biosphere's contribution from the fossil fuel CO<sub>2</sub> contribution derived from Δ<sup>14</sup>C and the total CO<sub>2</sub> enhancement over background, we can distinguish all three provided that there are large variations, such as in urban regions.





extraction, manometry, and mass spectrometry. Errors for  $\Delta^{14}\text{C}$  average 2‰, based on long-term reproducibility of secondary standards.

## 2.3 Calculations

A major goal of this study is the attribution of the sources of the  $\text{CO}_2$  emissions observed. A schematic figure of the flow of data used to calculate the portion of the total  $\text{CO}_2$  that is due to biosphere respiration (bio) and fossil fuel (ff) combustion, including burning of petroleum (pet) and natural gas (ng), is shown in Fig. 2. Mole fractions of  $\text{CO}_2$  measured at the two sites and a background site in La Jolla, CA, were used to calculate the  $\text{CO}_2$  excess (xs) over background (bg). The contributions of fossil fuel combustion and the biosphere to the excess were determined from radiocarbon measurements, and the fossil fuel component was further broken down into petroleum and natural gas using  $\delta^{13}\text{C}$  of the  $\text{CO}_2$ . Details are described below.

### 2.3.1 Total $\text{CO}_2$ emissions and background $\text{CO}_2$ mole fraction

The  $\text{CO}_2$  excess caused by local emissions at the two sites was calculated by subtracting an estimate of the background  $\text{CO}_2$  mole fraction derived from La Jolla monthly values (Keeling et al., 2005; Figs. 3 and 4). Flask sampling at La Jolla is done so as to minimize the influence of local  $\text{CO}_2$  sources by sampling during periods that simultaneously satisfy three criteria: low variability in  $\text{CO}_2$  concentration for periods of 3 h or more, wind speed of  $2.6 \text{ m s}^{-1}$  or more from a narrow southwesterly to westerly sector, and high visibility. That these methods successfully minimize influences of local fossil-fuel emissions is indicated by the consistency of the annual radiocarbon concentrations at La Jolla compared to clean stations both to the north and south in the Northern Hemisphere (Graven, 2012). In this paper, therefore, the La Jolla data presented are screened background data. The La Jolla data were interpolated to determine the appropriate value for the midpoint of the range of collection dates included in each  $\Delta^{14}\text{C}$  sample, using the algorithm from Thoning et al. (1989), with two harmonic terms, three

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polynomial terms, and the smoothed residuals of the long term trend (cutoff of 667 days).

### 2.3.2 CO<sub>2</sub> from fossil fuels, based on Δ<sup>14</sup>C

Mass balance calculations were used to calculate the relative contributions of background air, biosphere respiration and photosynthesis, and fossil fuel combustion (including natural gas and oil) to the CO<sub>2</sub> collected at the two sites. The following equations quantitatively separate the background air, biosphere, and fossil fuel combustion contributions to the locally measured atmospheric CO<sub>2</sub> using Δ<sup>14</sup>C (e.g., Levin et al., 2003; Miller et al., 2012; Pataki et al., 2003; Turnbull et al., 2006; Fig. 4):

$$C_{\text{obs}} = C_{\text{bg}} + C_{\text{ff}} + C_{\text{r}} + C_{\text{p}} \quad (1)$$

$$\Delta_{\text{obs}} C_{\text{obs}} = \Delta_{\text{bg}} C_{\text{bg}} + \Delta_{\text{ff}} C_{\text{ff}} + \Delta_{\text{r}} C_{\text{r}} + \Delta_{\text{p}} C_{\text{p}} \quad (2)$$

where subscripts obs, bg, ff, r and p indicate observed, background, fossil fuels, respiration, and photosynthesis, respectively,  $C$  indicates CO<sub>2</sub> mole fraction in ppm, and  $\Delta$  indicates Δ<sup>14</sup>C in ‰. We assume that Δ<sub>p</sub> is equivalent to Δ<sub>bg</sub> since natural fractionation during uptake is corrected in the Δ<sup>14</sup>C measurement and therefore substitute Δ<sub>bg</sub> for Δ<sub>p</sub> in Eq. (2). Then, after solving Eq. (1) for  $C_{\text{p}}$  and substituting this for  $C_{\text{p}}$  in Eq. (2), we solve Eq. (2) for  $C_{\text{ff}}$ , resulting in the following expression for  $C_{\text{ff}}$ :

$$C_{\text{ff}} = \frac{C_{\text{obs}} (\Delta_{\text{obs}} - \Delta_{\text{bg}})}{\Delta_{\text{ff}} - \Delta_{\text{bg}}} - \frac{C_{\text{r}} (\Delta_{\text{r}} - \Delta_{\text{bg}})}{\Delta_{\text{ff}} - \Delta_{\text{bg}}} \quad (3)$$

The value of Δ<sub>ff</sub> is −1000 ‰, since fossil fuels contain no <sup>14</sup>C because they have been removed from the source of this short-lived radionuclide for millions of years. We use the record from Pt. Barrow, AK (Xiaomei Xu, unpublished data) for the concurrent background Δ<sup>14</sup>C values (Δ<sub>bg</sub>), because these are the most complete record available for the entire time period of this study. Comparison with the data from La Jolla

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(Graven et al., 2012; Fig. 5) shows good agreement for 2004–2007, when the two data sets overlap. Comparing the calculated values for  $C_{ff}$  from these two backgrounds and propagating through the time series calculations (Sect. 3.4) results in a difference of approximately 1 % of the signal we are measuring. We calculate  $C_{bio}$  (the sum of  $C_r$  and  $C_p$ ) from Eq. (1), using the calculated values of  $C_{ff}$  and the independent estimates of  $C_{bg}$  from the La Jolla data, so that we understand the contribution of the biosphere to total local emissions. The nuclear power plant contribution, the only other source of  $^{14}C$ , is small on the west coast of the US (Graven and Gruber, 2011).

Following Turnbull et al. (2006) and Miller et al. (2012), the respiration terms in the equations above are assumed to reflect contributions due to heterotrophic respiration. Thus, the second term in Eq. (3) is small in magnitude and is due to heterotrophic respiration, through which microbes respire  $CO_2$  that was from carbon previously incorporated through photosynthesis. This term takes into account the isotopic disequilibrium due to the significant time delay between photosynthetic incorporation and respiration, assumed to be 10 years on average (Miller et al., 2012). The magnitude of this correction for our urban Pasadena site is different relative to sites with smaller anthropogenic  $CO_2$  signals, since the  $CO_2$  photosynthesized into the plant a decade ago was not close to the background air composition of that time but was the local, “polluted” air. The  $\Delta_r$  in Eq. (3) for each sample was calculated by extrapolating the Pasadena trend back 10 years. Because of the mild climate in southern California, we used a constant value of  $C_r = 5$  ppm, the same value used for summer by Turnbull et al. (2006). This should be taken as an upper limit for this urban region. The range of the correction for the second term in Eq. (3), including the sign, was  $-0.06$  to  $-0.11$  ppm, generally smaller relative to regions where the biosphere contribution  $C_r$  is large (Miller et al., 2012; Turnbull et al., 2006). For the data from the Palos Verdes site, we calculated the heterotrophic correction term using values of  $\Delta_r$  calculated by extrapolating the Pt. Barrow background trend back 10 years and used a constant value of  $C_r = 5$  ppm, because of the mild climate. The correction term for the Palos Verdes data ranged from



the eight-year sampling period in Pasadena, were excluded since they fell more than three times the standard error from their linear regression best-fit lines. The monthly MT plots for 2011 are shown in Fig. A1, as examples.

We use the results from the  $^{14}\text{CO}_2$  calculations for the fraction of  $C_{\text{xs}}$  from the biosphere ( $F_{\text{bio}} = 1 - F_{\text{ff}}$ ) together with the MT slopes to attribute the  $\text{CO}_2$  derived from petroleum and natural gas combustion ( $C_{\text{pet}}$  and  $C_{\text{ng}}$ ) by mass balance, first by calculating the  $\delta^{13}\text{C}$  of the fossil fuel component, using:

$$\delta_{\text{ff}} = \frac{\delta_{\text{xs}} - \delta_{\text{bio}} \times (1 - F_{\text{ff}})}{F_{\text{ff}}} \quad (7)$$

where  $F_{\text{ff}}$  is the fraction of  $C_{\text{xs}}$  due to emissions from fossil fuel combustion, as calculated from the  $^{14}\text{CO}_2$  data. The values for  $\delta_{\text{xs}}$  are the seasonal  $\delta^{13}\text{C}$  values from the MT analyses and  $\delta_{\text{bio}}$  is taken to be  $-26.6\text{‰}$ , the average  $\delta^{13}\text{C}$  of the ambient air plus the discrimination of  $-16.8\text{‰}$  for the biosphere (Bakwin et al., 1998). Then the proportions of  $\text{CO}_2$  emitted by petroleum and natural gas combustion are calculated using the  $\delta^{13}\text{C}$  values:

$$\delta_{\text{ff}} = F_{\text{petff}} \times \delta_{\text{pet}} + (1 - F_{\text{petff}}) \times \delta_{\text{ng}} \quad (8)$$

$$F_{\text{petff}} = \frac{\delta_{\text{ff}} - \delta_{\text{ng}}}{\delta_{\text{pet}} - \delta_{\text{ng}}} \quad (9)$$

with an analogous equation for  $F_{\text{ngff}}$  where  $F_{\text{petff}}$  and  $F_{\text{ngff}}$  are the fractions of petroleum and natural gas combustion contributions in  $C_{\text{ff}}$ , respectively. The values of  $\delta_{\text{ng}}$  and  $\delta_{\text{pet}}$  used were  $-40.2\text{‰}$  for natural gas (Newman et al., 2008) and  $-25.5\text{‰}$  for petroleum combustion (average of measurements in Newman et al., 2008, and  $-25.1\text{‰}$  measured in 2012). The  $C_{\text{ff}}$ ,  $C_{\text{pet}}$ , and  $C_{\text{bio}}$  components were corrected for the presence of 10% ethanol in California gasoline by multiplying  $C_{\text{pet}}$  by 0.067 (the fraction of  $\text{CO}_2$  emitted by burning the ethanol portion of the ethanol-gasoline mixture; EIA, 2015) to

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its 8 year record. Then we compare the results with government inventories and with the high-resolution Hestia-LA emissions product.

### 3.1 Spatial variations – comparison of source attribution at the Pasadena and Palos Verdes sites

5 The  $\Delta^{14}\text{C}$  time series for the two sites are shown in Fig. 3c and d, 8 years for Pasadena and 4 years for Palos Verdes. The two data sets are very different, with Palos Verdes radiocarbon results being significantly higher than those in Pasadena except during the winter. The summer months in Pasadena are characterized by  $\Delta^{14}\text{C}$  values far from background, i.e., depleted in  $^{14}\text{C}$  due to dilution by  $\text{CO}_2$  produced by burning of  
10 fossil fuels containing none of the radioactive isotope. There are occasional negative spikes in  $\Delta^{14}\text{C}$  during the winter. Total  $\text{CO}_2$  excess ( $C_{\text{xs}}$ ; Fig. 4), determined as  $\text{CO}_2$  concentration minus background, is similarly disparate with respect to timing. The total enhancement at both Pasadena and Palos Verdes,  $C_{\text{xs}}$ , spikes during winter (up to 65 and 34 ppm, respectively), but the Pasadena excess also peaks during the summer (up to 43 ppm), whereas Palos Verdes values for  $C_{\text{xs}}$  are at a minimum during the warm  
15 months (3–20 ppm). When the  $^{14}\text{C}$  and  $C_{\text{xs}}$  information are combined to calculate  $\text{CO}_2$  emissions due to fossil fuels ( $C_{\text{ff}}$ ; Eq. 3; Fig. 4), we see summer maxima for  $C_{\text{ff}}$  in Pasadena, but not in Palos Verdes. The spikes in  $C_{\text{xs}}$  and  $C_{\text{ff}}$  during fall and winter seasons are not the general trend in Pasadena, as evidenced by the quarterly averages (Fig. 6b). The amount of  $C_{\text{ff}}$  in the Pasadena seasonal averages (Figs. 4a and 6b) ranges from  $(18.9 \pm 1.2)$  ppm (winter) to  $(26.8 \pm 0.4)$  ppm (summer). In Palos Verdes,  $C_{\text{ff}}$  averages  $(5 \pm 3)$  ppm during the warmer months and  $(12 \pm 5)$  ppm during the winter months (Fig. 4b). However,  $\text{CO}_2$  emissions from the biosphere ( $C_{\text{bio}}$ ) tend to be higher during the cooler months at both sites (Fig. 4).

25 The explanation for the differences in the seasonal cycles of  $C_{\text{xs}}$  and  $C_{\text{ff}}$  at the two sites is probably the different wind patterns for the different times of year. Figure 7 shows back trajectories ending at 14:00 PST in Pasadena (Fig. A2 for both sites), calculated using NOAA's HYSPLIT model (Draxler and Rolph, 2014.), for January and

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July 2011. These are representative of these months in all years of this study. Wind directions during July are from the west-southwest, whereas they are mostly from the northeast but much more varied during the winter. Thus, in Pasadena, elevated  $C_{xs}$  and  $C_{ff}$  values during the summer result from air masses traveling across the Los Angeles basin, picking up emissions and transporting them inland. During the winter, the airflow is more mixed, resulting in lower average  $C_{ff}$  signals in Pasadena, since a significant proportion of the winds bring less polluted air from the much less populated mountains and deserts located to the north (Santa Ana winds) (Fig. 7). The summer westerly winds bring ocean air to the Palos Verdes site, characterized by  $CO_2$  mole fractions and  $\Delta^{14}C$  very similar to background marine air. During the cooler months, the Santa Ana winds from the northeast occasionally blow over the LA basin, bringing its emissions to the coastal site (Fig. 7). This pattern results in more scatter in the magnitude of  $CO_2$  excess observed during the winter at the Palos Verdes site, than during the summer. Figure 8 shows the average annual patterns for  $C_{ff}$  at the two sites, demonstrating the effect of the varying wind direction patterns.

### 3.2 Attribution of $CO_2$ excess from different anthropogenic sources for Pasadena

Since we have information regarding the relative contributions of fossil fuel combustion and biosphere respiration from the radiocarbon data, we can use the differences in the  $\delta^{13}C$  of the  $CO_2$  to look at the contributions of petroleum/gasoline vs. natural gas combustion. We use the MT approach to distinguish between different fossil fuel sources of  $CO_2$  (Miller and Tans, 2003). As described in Sect. 2.3.3, the MT slope of the correlation gives the  $\delta^{13}C$  of the local source of  $CO_2$  emissions. In many cases it is difficult to distinguish the anthropogenic sources because the biosphere's signal can overlap that of petroleum. However, in a megacity such as the Los Angeles basin, the contribution of the biosphere to the total  $CO_2$  enhancement can be minimal ( $\leq 20\%$  in Pasadena; Newman et al., 2008, 2013) during the afternoon, when the boundary

layer is deepest and most thoroughly mixed. In this study, we use the information from  $\Delta^{14}\text{C}$  presented above to further constrain the biosphere's input. Since the other major anthropogenic sources (cement production and combustion of coal) are not present in the Los Angeles basin,  $\delta^{13}\text{C}$  from MT plots can be used to differentiate the proportions of natural gas and oil burned in the region, as discussed below.

Seasonal MT slopes for the mid-afternoon Pasadena samples from 2006 through 2013 are shown in Fig. 6a. We do not present similar analysis for the Palos Verdes data because it is a shorter data set, with only 3–5 measurements per month (12 per season), and the range in  $\text{CO}_2$  mole fractions during the warmer months is less than 20 ppm for all spring and summer seasons. Thus there are insufficient meaningful data to produce a significant trend. Vardag et al. (2015) came to this same conclusion for a rural site in Germany, based on a modeling study.

The  $\delta^{13}\text{C}$  values from MT regressions for the cooler portions of the year in Pasadena are almost always higher than those for the warmer portions. The values for the cooler seasons average  $(-30.6 \pm 0.5)\text{‰}$ , 1.8‰ higher than the average for the warmer months,  $(-32.4 \pm 0.6)\text{‰}$ . Newman et al. (2008) showed that no biospheric input was required to explain the  $\delta^{13}\text{C}$  of mid-afternoon  $\text{CO}_2$  during 1972–1973 and 2002–2003, although up to 20% was allowed by the uncertainties. Assuming that there is no contribution from respiration and that the  $\delta^{13}\text{C}$  of the high- $\text{CO}_2$  end members are  $-40.2\text{‰}$  for natural gas and  $-25.5\text{‰}$  for petroleum combustion, as discussed above, then the proportion of natural gas burned in  $C_{\text{XS}}$  is 32% during the cooler months and 45% during the warmer months. The larger fraction of natural gas burned during the warm part of the year is consistent with the observed burning of more natural gas for electricity generation during summer months, as would be required to power air conditioning needs. Mild winters in this climate require less natural gas combustion for heating buildings, thus minimizing a large winter peak frequently seen in colder regions, such as Salt Lake City, UT (Pataki et al., 2003; Bush et al., 2007) and Chicago, IL (Moore and Jacobson, 2015).

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analysis of Jiang et al. (2008; Fig. 9b and h). The sum of the seasonal and semi-annual harmonic terms reproduces the data very well, with  $R^2$  values of 0.70 and 0.91 for Pasadena and background, respectively. The average annual cycles are 6 months out of phase, whereas the semi-annual oscillation cycles look very similar at the two sites.

The seasonal cycle in Pasadena is consistent with influx of combustion CO<sub>2</sub> during the hot summer months due to increased burning of natural gas at power plants located dominantly in the southwestern portion of the LA basin (CEC, 2015). In contrast, the background data reflect global patterns with a drawdown in CO<sub>2</sub> during the summer growing season in the Northern Hemisphere. Jiang et al. (2012) concluded that the semi-annual oscillation at NOAA's GLOBAL-VIEW sites is due to the combination of net primary production and respiration of the biosphere. The semi-annual oscillation in the background signal is consistent with this interpretation. We see virtually the same pattern in Pasadena, although the amplitude is smaller, consistent with the small biospheric contribution indicated by the  $\Delta^{14}\text{C}$  results.

Based on the work of Jiang et al. (2012) we expect the annual cycle in Pasadena to be larger in amplitude than in La Jolla since it is further north, but the amplitude is actually much smaller. If the regional emissions of CO<sub>2</sub> in Pasadena are relative to a La Jolla background, then there is a huge enhancement during the summer! Indeed, the seasonal cycle for  $C_{\text{ff}}$  (Fig. 8) is 11 ppm, with the peak in August–September, and there is very little semi-annual oscillation.

The annual pattern for CO<sub>2</sub> in Palos Verdes is also heavily influenced by the transport of combustion emissions from the Los Angeles basin (Figs. 7, 8, 9c and d). The average monthly pattern is more similar to the background's (Fig. 9g and h) than to Pasadena's (Fig. 9a and b). However, there is a strong peak in the winter that is consistent with the increased number of days during this time of year with winds from the north to east, travelling over the basin. Doing the same analysis for the monthly minimum values (Fig. 9e and f) gives a pattern that is much more similar to the background's, confirmed by the comparison of the raw data with the background smoothed time series in Fig. 3. This supports use of minimum values from Palos Verdes as reasonable background



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(Fig. 10f). The sum of the trend + IMF 6 is a curve with increasing  $C_{ff}$  values leading up to mid-2007, when they began to fall, until leveling off in 2010 and perhaps starting to rise towards the end of the time series. There are end effects in this method, such that we do not have confidence in the first and last years of the analysis. The uncertainties in this calculation are shown by the shaded regions in Fig. 10f. These were determined as the  $1\sigma$  standard deviations of adding random noise equivalent to 13.7 % to the data 300 times and then running the EMD analysis. The 13.7 % noise added is the uncertainty of the  $C_{ff}$  values calculated from  $\Delta^{14}C$ ,  $\pm 1$  ppm, relative to the standard deviation of the data, 7.3 ppm. The maximum and minimum values are distinct at approximately the  $2\sigma$  standard deviation level, as shown in Fig. 10f and indicate a significant decrease of 9.5 % between the maximum in May 2007 (average from Fig. 10a plus the maximum in Fig. 10f: 23.7 ppm) and the average for January–June 2010 (average from Fig. 10a plus the average of minimum six-month period from Fig. 10f: 21.4 ppm). Using different backgrounds for  $\Delta^{14}C$ , such as extrapolating the data from La Jolla (Fig. 5) does not significantly affect this analysis, resulting in differences of  $(0.01 \pm 0.09)$  ppm  $C_{ff}$  out of a range on the order of 2 ppm. And our result showing that there are different values of  $\Delta^{14}C$  for bulk fuel for autumn–winter than for spring–summer also does not change these conclusions, since the RMSE of the IMF6+ trend (Fig. 10f) using different  $\Delta^{14}C$  for cool vs. summer months relative to the constant average value is 0.1 ppm  $C_{ff}$ .

The timing of the drop in the fossil-fuel CO<sub>2</sub> excess around 2008 is consistent with the economic recession in late 2007–2009 (NBER, 2010) with slow recovery beginning in 2010. Similar results for global CO<sub>2</sub> emissions due to fossil fuel combustion have been documented by Peters et al. (2012) and Asefi-Najafabady et al. (2014). The fraction of decrease in  $C_{ff}$  (9.5 %) is similar to, although less than, the decrease in global GDP during this time (global GDP decreased by 13 %; World Bank, 2015). The observation that  $C_{ff}$  does not decrease by as large a percentage as the economy is consistent with the analysis of York (2012) that anthropogenic emissions are asymmetric with respect to economic growth and decline. Increases in emissions that come during growth periods are not matched by declines of the same magnitude during economic downturns, be-



tion data, based on deliveries (EIA), and gasoline taxes collected (CBE, 2014) with  $C_{\text{pet}}$  indicates similar decreases of 10–20 %, but with a lag of a few months (Fig. 12). The EIA and State of California data reflect the entire state domain while the Hestia emissions data product reflects the LA Basin specifically. Furthermore, the EIA data and the gasoline tax-based data will likely be displaced in space and time from the point of combustion, which is dominated by on-road vehicles. The Hestia data, by contrast, is a fossil fuel  $\text{CO}_2$  emissions data product specific in space and time to the individual building and road segments (Gurney et al., 2012).

To truly understand the observations in Pasadena, we must combine information from spatial and temporal meteorological and emissions databases, such as obtained using a model like the Weather Research and Forecasting (WRF) model. Since this is beyond the scope of this work, we have used the information from HYSPLIT back trajectories (Fig. 7; January and July) to provide rough limits for winds arriving in Pasadena at our sampling time of 14:00 PST. These back trajectories suggest that prevailing winds during the summer come from the southwest, across the basin, and winds during the winter come from the northeast, across the mountains from the desert. We have looked at  $1.3\text{ km} \times 1.3\text{ km}$  gridded emissions from the Hestia-LA data product to qualitatively determine what relative emissions from petroleum combustion are expected during January and July for the two years of the Hestia data (2011 and 2012). These are plotted in Fig. 12 and agree with the observations presented here, that more  $C_{\text{pet}}$  is observed during the summer than during the winter. A map of the regions selected for January (NE) and July (SW) is presented in Fig. 13a, along with the HYSPLIT back trajectories for January and July 2011, and the monthly average  $\text{CO}_2$  emissions due to total petroleum combustion (the Hestia-LA product) from the two integrated areas based on the wind directions are shown in Fig. 13b for years 2011 and 2012.

We show comparison of the  $C_{\text{ng}}$  results from Pasadena with area-integrated bottom-up inventories and the Hestia-LA data product in Fig. 14. The California Energy Commission (CEC, 2015) compiles data for natural gas consumed by power plants throughout the state, including Los Angeles and Orange counties. These seasonal data are

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consistent with the detailed Hestia-LA data for the electricity production for the entire Los Angeles basin (dashed dark blue line in Fig. 14a). And the seasonality of all of the inventories involving just the electrical power sector agrees well with the seasonality of the time series for  $C_{ng}$  (Fig. 14a), with peaks during the summer and troughs during the winter. The source attribution analysis using  $\Delta^{14}C$  and  $\delta^{13}C$  also captures the increase in  $C_{ng}$  consumption of the power plants in recent years, although the data from this study suggest that the increase started earlier than do the inventories. However, the observations of  $CO_2$  concentration and  $\delta^{13}C$  integrate over all natural gas combustion and cannot pick out just this one sector.

Overall statewide and Los Angeles basin inventories show maximum natural gas usage during the winter (dashed green line in Fig. 14b). Other sources of combusted natural gas include residential, commercial, industrial, and transportation use, which could affect the trends, but we do not have seasonal data for these in the Los Angeles megacity for the full period of this study. However, the seasonal signal for total emissions from natural gas combustion from the Hestia-LA project for 2011–2012 is consistent with the data presented here, when the seasonal prevailing wind directions are considered. The seasonal pattern of emissions from natural gas combustion at any one location is characterized by a small peak during the cooler months and a trough during the warmer months (Fig. 13c). However, emissions in the region sampled by winds arriving in Pasadena during the winter (the northeast) are always lower than those in the basin, over which the summer winds travel to the sampling site. Therefore, transport of air masses following the seasonal wind patterns can explain the observations in Pasadena.

Since the seasonal cycle observed in  $C_{pet}$  and  $C_{ng}$  in Pasadena is probably due to atmospheric transport, modeling of this effect is critical to being able to combine top-down observations and bottom-up economic and usage data for a direct consistency comparison. These effects must be removed in order to understand long-term trends due to variations in anthropogenic emissions. The time series analysis using Empirical Mode Decomposition presented in Sect. 3.4.1 removes the seasonal signals to

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concentrate on the longer-term signals, which show reasonable agreement with the longer-term trends in the statewide inventory.

#### 4 Conclusion

Detection of anthropogenic excess of CO<sub>2</sub> at two sites in the Los Angeles basin, one on the coast and one inland against a barrier mountain range, reveals significant spatial and seasonal variability due to the biosphere, natural gas combustion, and petroleum combustion. Seasonal patterns in wind direction determine the source region of the excess detected at the two sites. Winds from the west to southwest during the warmer months bring marine air with little excess to Palos Verdes, and these same winds continue across the LA basin picking up emissions from fossil fuel combustion to be observed in Pasadena. During the cooler months, wind directions are more varied and include periods when air with low emissions comes to Pasadena from the northeast to northwest and then travels across the basin to Palos Verdes, incorporating anthropogenic emissions along the way.

The nature of the excess changes with season, as reflected by the  $\delta^{13}\text{C}$  values of CO<sub>2</sub> observed in Pasadena. During warmer months, lower values for  $\delta^{13}\text{C}$  of the local excess indicate a higher proportion of natural gas burned, consistent with government inventories that indicate more natural gas burned during summer to produce electricity to power air conditioning. Even more importantly, however, the seasonal trends in the fossil fuel combustion observed in Pasadena are consistent with the shift from southwesterly winds during warmer months to northeasterly winds during cooler months. Therefore the source region of emissions changes from the Los Angeles basin during summer to the mountains and desert during winter, for our Pasadena sampling site. Trend analysis by ensemble empirical mode decomposition supports the relationship between emissions and temperature.

The long-term trend in CO<sub>2</sub> excess from fossil fuel combustion is consistent with emissions changes associated with the economic recession and slow recovery of 2008

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through the present, and indicates a significant decrease of 9.5 % since the maximum in late 2007, consistent with the bottom-up inventory of the California Air Resources Board. Indeed, top-down and bottom-up methods of determining the anthropogenic sources of CO<sub>2</sub> emissions must be compared to each other to better understand inconsistencies, potential biases, and uncertainty. Previously, however, comparisons have been limited by the scope of emissions, large and overlapping uncertainty, and differences in the target domain. Here we have shown that combining data from radiocarbon and  $\delta^{13}\text{C}$  values moves us towards a direct comparison in a megacity with very large emissions. Measurement trends at a receptor site are consistent with annual variations in California statewide bottom-up inventories for CO<sub>2</sub> emissions attributed to petroleum and natural combustion, individually as well as for total CO<sub>2</sub> emissions. Even greater consistency between top-down measurements and granular emission estimates specific for the LA megacity domain are achieved when considering wind direction and sub-city source regions. This strengthens the need to have measurement, modeling, and inventories that are specifically aimed at the same domain with fine space/time resolution.

The next steps are to include modeling with inversion of the measurements to understand the combination of atmospheric transport and emissions and to extend the analysis to a denser network of surface monitoring stations such as the Los Angeles Megacities Carbon Monitoring Project (Kort et al., 2013) and the California Laboratory for Atmospheric Remote Sensing (CLARS) observations from Mount Wilson (Wong et al., 2015). Although the uncertainties are large enough that the method described here will not be usable in non-urban regions, similar to the conclusion of the modeling study by Vardag et al. (2015), anthropogenic emissions dominate significantly over natural processes in megacities. Therefore, this kind of monitoring in megacities will allow society to understand and monitor the sources of the CO<sub>2</sub> that are the major contributors to global warming.

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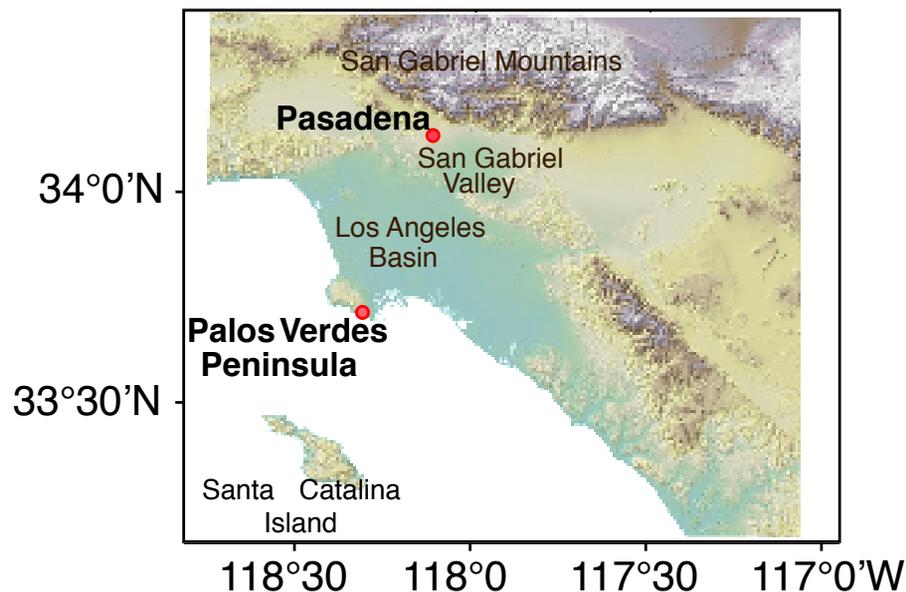
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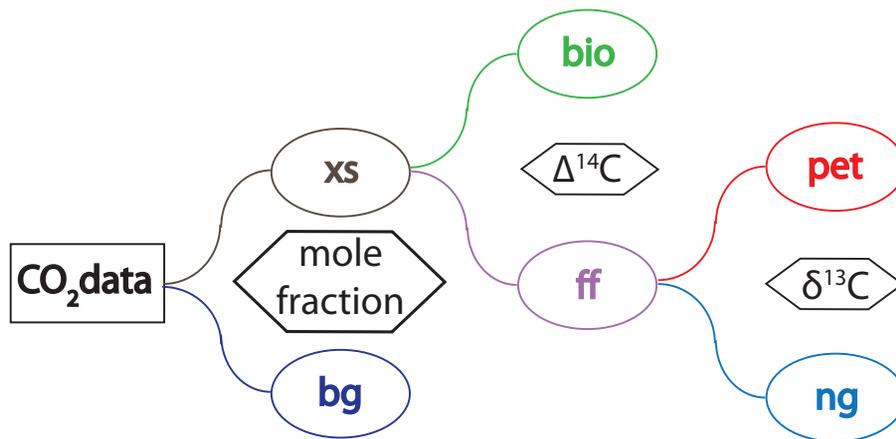
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**Figure 1.** Map of southern California, showing sampling locations in Pasadena and Palos Verdes (red dots).

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**Figure 2.** Schematic diagram showing the use of different data sets for attribution of the sources of CO<sub>2</sub> emissions. Mole fractions of background (bg) and observations are used to determine  $C_{xs}$  (excess over background/bg);  $\Delta^{14}\text{C}$  values are used to distinguish  $C_{ff}$  (fossil fuel, ff) and  $C_{bio}$  (biosphere, bio);  $\delta^{13}\text{C}$  compositions are used to distinguish  $C_{pet}$  (petroleum/gasoline, pet) from  $C_{ng}$  (natural gas, ng).

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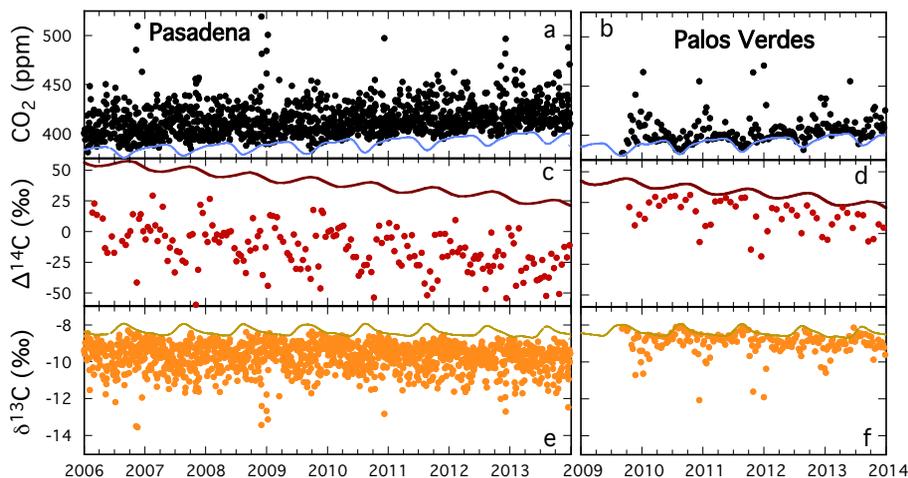
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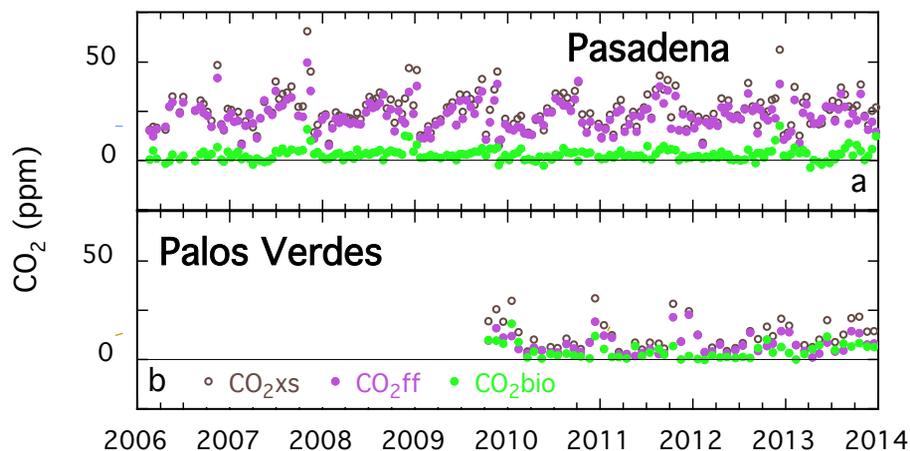


**Figure 3.** Time series of CO<sub>2</sub> mole fractions for <sup>14</sup>CO<sub>2</sub> samples (a, b), Δ<sup>14</sup>C data (c, d), and δ<sup>13</sup>C (e, f) for Pasadena and Palos Verdes. The solid curves are backgrounds used in the calculations: δ<sup>13</sup>C and CO<sub>2</sub> backgrounds are from La Jolla, CA and Δ<sup>14</sup>C from Pt. Barrow, AK.

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**Figure 4.** Time series of  $C_{\text{XS}}$ ,  $C_{\text{ff}}$ , and  $C_{\text{bio}}$  calculated from  $\Delta^{14}\text{C}$  (see text for description of calculations) for Pasadena (top) and Palos Verdes (bottom). The errors for  $C_{\text{ff}}$  are 1 ppm. The negative  $C_{\text{bio}}$  values indicate photosynthetic uptake. The value of  $\Delta^{14}\text{C}$  for fuel for this calculation was taken to be  $-954\text{‰}$ , the average from the summer and winter calculations.

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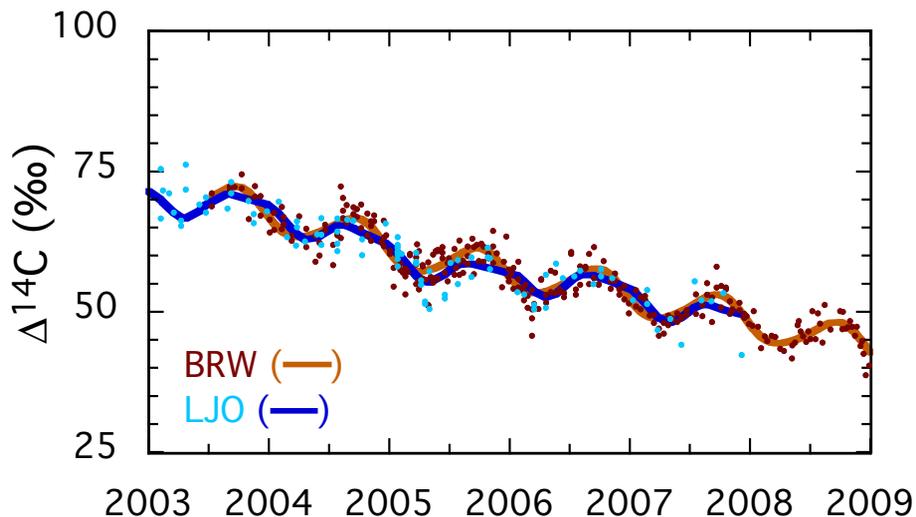
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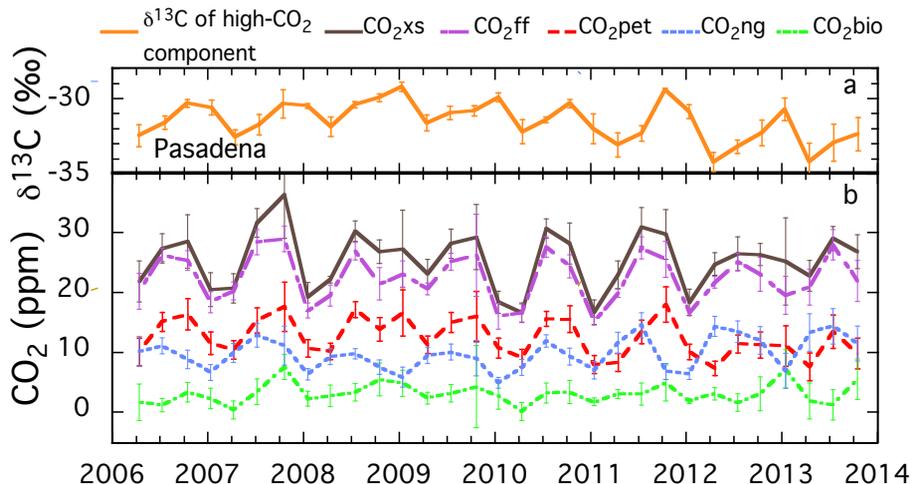


**Figure 5.** Comparison of possible background records for this study, Pt. Barrow, AK, (BRW; Xiaomei Xu, unpublished data) and La Jolla, CA (LJO; Graven et al., 2012). The smoothed brown curve for BRW is the  $\Delta^{14}\text{C}$  background used for this study and was calculated using the algorithm of Thoning et al. (1989), from the function plus the smoothed residuals of the long-term trend, using 2 harmonic and 3 polynomial terms in the function and 667 days as the long-term cutoff for the low-pass filter.

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**Figure 6.** Attribution of CO<sub>2</sub> excess in Pasadena among combustion of natural gas and petroleum and the biosphere. **(a)** Miller–Tans slopes for seasonal averages of monthly plots. Error bars are standard errors of the regression intercepts. **(b)** Attribution of C<sub>xs</sub> among all three sources (natural gas, petroleum, and the biosphere), combining the information from  $\Delta^{14}\text{C}$  and  $\delta^{13}\text{C}$ , using Miller–Tans slopes to determine the relative proportions of petroleum and natural gas combustion. Error bars are propagated from the errors in the  $\delta^{13}\text{C}$  intercepts and the  $\Delta^{14}\text{C}$  measurements.

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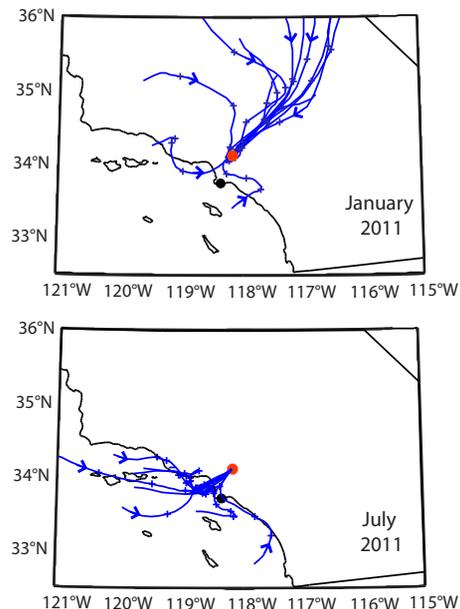
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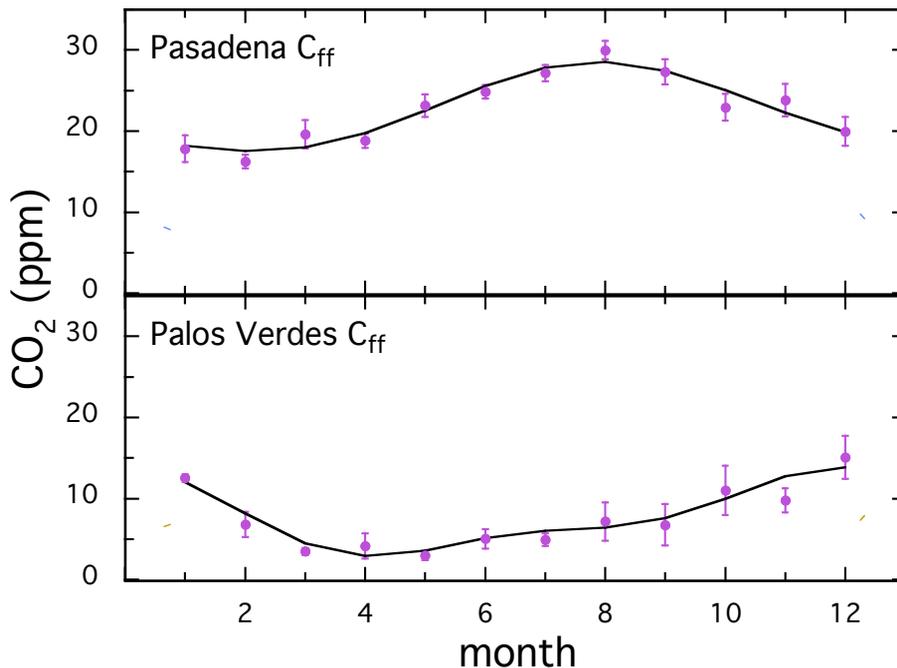


**Figure 7.** Back trajectories (24 h) for winds arriving at the Pasadena site (red dot) at 14:00 PST for January **(a)** and July **(b)** 2011, calculated by HYSPLIT (Draxler and Rolph, 2015) for all sampling days in January and selected sampling days in July, for clarity. Results for all sampling days are shown in Fig. A2. Arrows indicate the direction of air flow. Plus signs indicate 6, 12, and 18 h from the Pasadena site. The black dot is the location of the Palos Verdes site. The back trajectories for the Palos Verdes site show a similar pattern (Appendix Fig. A2). The back trajectories explain the difference between the annual cycles at the two sites, shown in Fig. 8.

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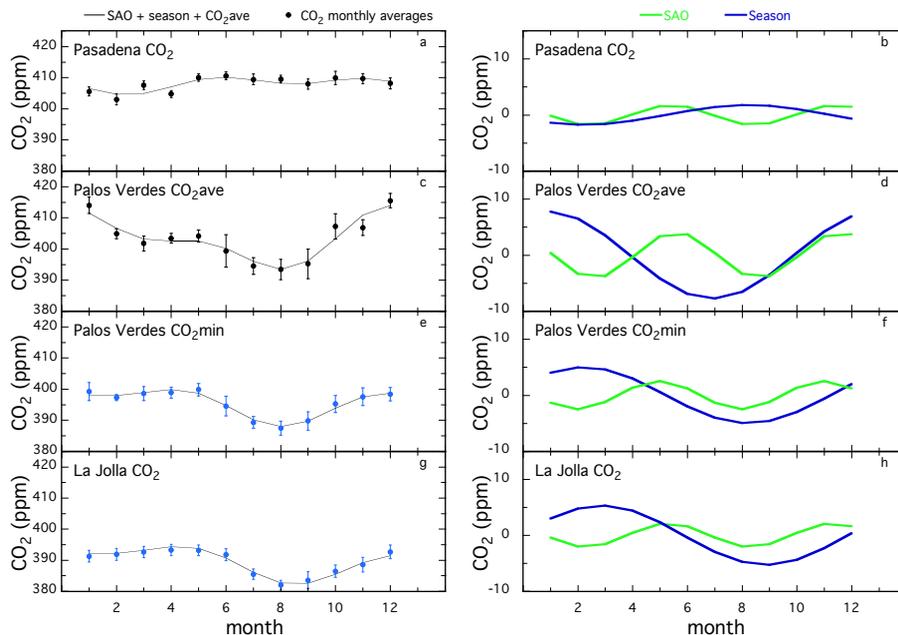


**Figure 8.** The annual patterns for  $C_{ff}$  in Pasadena and Palos Verdes calculated as the best fit of two harmonics plus the average of the annual cycles (black curves). These patterns are consistent with seasonal differences in the back trajectories shown in Fig. 7.

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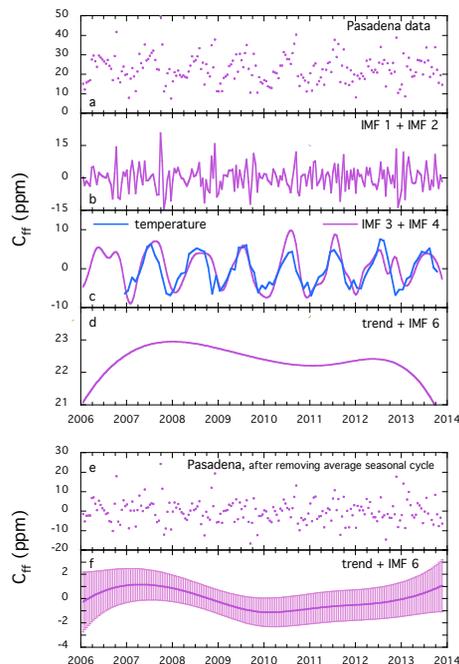
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**Figure 9.** Comparison of seasonal and semi-annual oscillation cycles of CO<sub>2</sub> mole fractions for flask samples from Pasadena (2006–2013) with those at the La Jolla (2006–2013) and Palos Verdes (2009–2013) sites. Left column panels show the average annual patterns for the monthly averages together with the sum of the harmonics for seasonal (blue) and semi-annual (green) cycles (Jiang et al., 2008). Right column panels show the amplitudes and phases of the pure harmonic components. Two sets of results are shown for Palos Verdes, for the monthly averages (**c**, **d**) and for the monthly averages of weekly minima (**e**, **f**). The monthly averages show the effect of transport on the signal, with a large peak during the winter, while the minima (in blue) show that data from this site are very similar to La Jolla (in blue) and should be a good estimation of the background air for the LA basin. Error bars on the monthly averages of the data are  $1\sigma$  standard errors.

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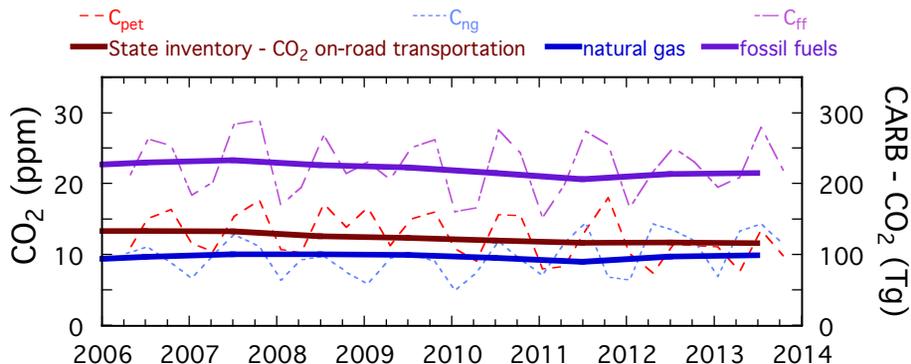
**Figure 10.** Results of ensemble empirical mode decomposition (EEMD) (Huang et al., 1998; Wu and Huang, 2009) of the  $C_{ff}$  time series calculated using Eq. (3) and the average, constant  $\Delta^{14}\text{C}$  of  $-954\text{‰}$  for fossil fuel. The top set of panels show the raw data (**a**), noise (**b**), annual and semi-annual mode (**c**), and the trend + IMF 6 (**d**). The pattern of the trend + IMF 6 shown in (**d**) is within  $1\sigma$  uncertainty of no variation over this time period. The bottom two panels include the raw data after subtracting the average annual cycle (**e**), and the trend + IMF 6 for the modified data set (**f**). 30 day average temperatures (minus the overall average and scaled to match the magnitude of the  $C_{ff}$  IMF; blue curve) are superimposed on the plot of IMF 3 + IMF 4 (**c**). Shaded regions in (**f**) indicate  $1\sigma$  standard deviation of 300 Monte Carlo realizations with 13.7 % noise added, the ratio of the uncertainty in  $C_{ff}$  to the standard deviation of the data.

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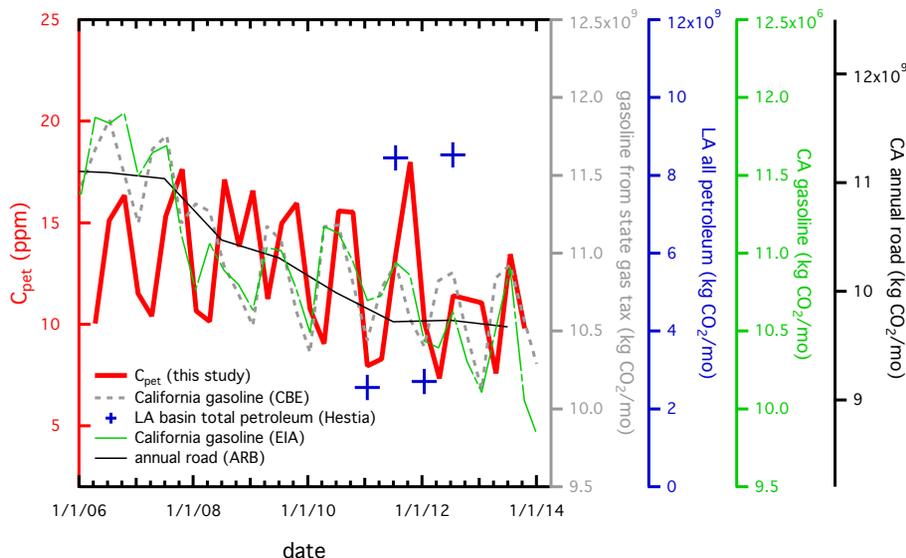
**Figure 11.** Comparison of annual average CO<sub>2</sub> emissions from bottom-up California Air Resources Board (CARB) inventories (thick lines; right axis labels) for fossil fuel-derived emissions with top-down seasonal averages from the Pasadena data, using the Miller and Tans (2003) approach to attribute CO<sub>2</sub> emissions from petroleum and natural gas combustion from the  $\delta^{13}\text{C}$  measurements. Seasonal curves showing the attribution of  $C_{\text{xs}}$  from Fig. 6b are shown as thin lines. The annual trends from the bottom-up CARB inventories are plotted on a scale exactly 100 times that of the trends derived from the CO<sub>2</sub> measurements, showing that the relative proportions are very similar through 2013.

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**Figure 12.** Comparison of the Pasadena  $C_{pet}$  atmospheric concentration with all available area-integrated bottom-up fossil fuel CO<sub>2</sub> emissions, including gasoline sales based on taxes paid to the California Board of Equalization (CBE, 2014), gasoline provided in California by prime suppliers, the California Air Resources Board’s annual road emissions (CARB, 2015), and the Hestia-LA gridded total petroleum. The Hestia-LA data product is specific to the Los Angeles megacity domain; all inventories are statewide estimates. Since the Hestia-LA product is gridded, we show the emissions emanating from different regions for January (northeast quadrant, Fig. 13a) and July (southwest quadrant), based on prevailing winds during those periods (Figs. 7 and 13a). The axis for each inventory has been adjusted to allow easy comparison. The seasonality of the  $C_{pet}$  data lags the bottom-up inventories by a few months. This analysis is consistent with the observed decrease in gasoline combustion during 2008–2011.

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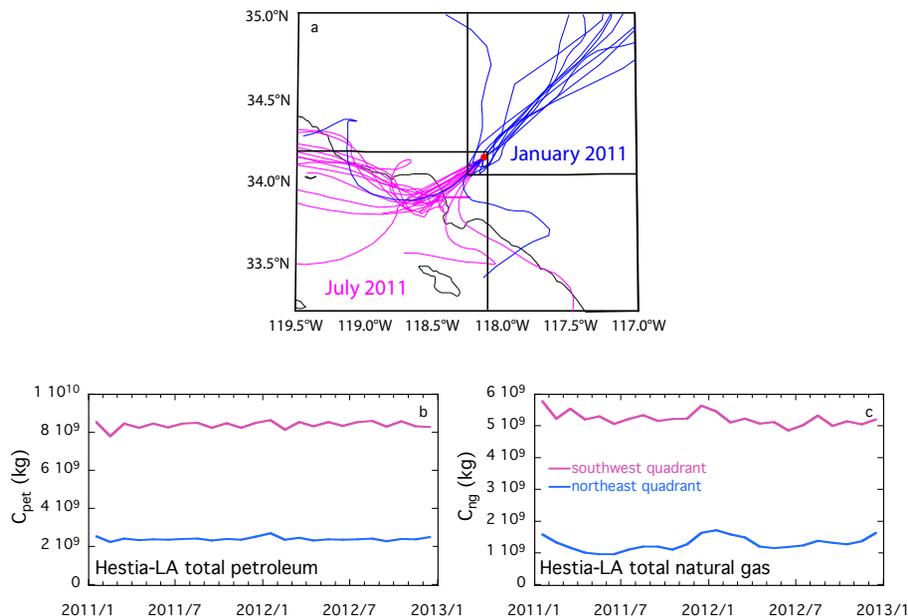
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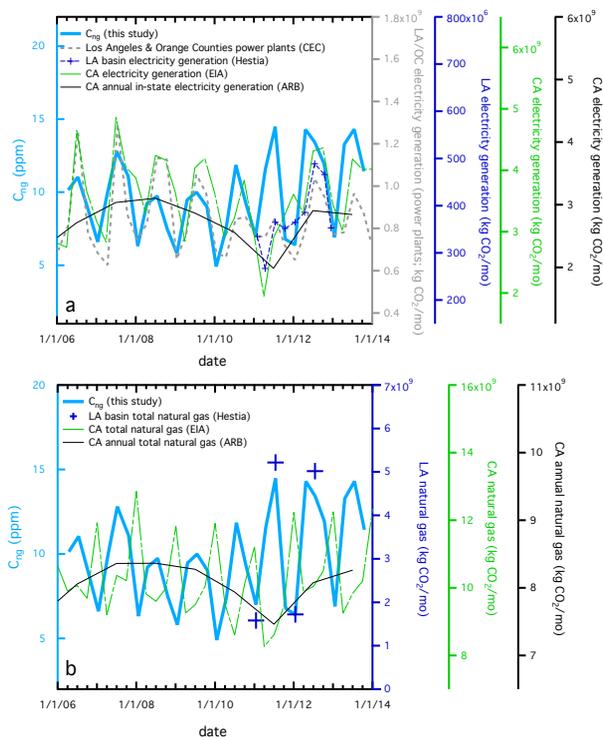
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**Figure 13.** Relevant emissions selection from the Hestia-LA data product. **(a)** Quadrants selected for investigation of CO<sub>2</sub> emissions from the Hestia-LA data product, together with the 24 h back trajectories calculated by HYSPLIT for January (northeast quadrant) and July (southwest quadrant)  $\Delta^{14}\text{C}$  sampling days. The back trajectories end in Pasadena (red dot) at 14:00 PST. Monthly averaged time series for Hestia-LA data product  $C_{\text{ff}}$  are shown from total petroleum combustion **(b)** and total natural gas combustion **(c)** for 2011 and 2012. For both the northeast quadrant of the Los Angeles region, the source of winter emissions, and the southwest quadrant, the source of summer emissions, the seasonal pattern is either flat (petroleum) or characterized by peaks during the winter (natural gas). But the summer emissions are always higher than those during winter, consistent with the observed top-down patterns for  $C_{\text{pet}}$  and  $C_{\text{ng}}$  in Pasadena.

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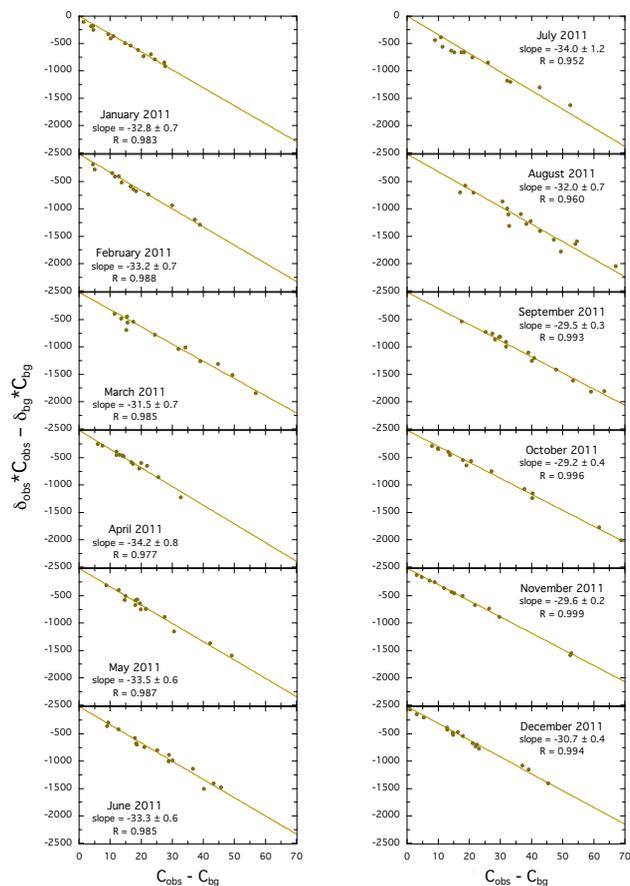
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**Figure 14.** Comparison of Pasadena  $C_{ng}$  atmospheric concentration with area-integrated inventories of natural gas combustion, as well as the gridded Hestia-LA data product for southwest and northeast regional sectors for July and January months, respectively. Panel (a) compares the data from this paper with usage of natural gas by the electrical power sector; panel (b) shows the comparison with total natural gas consumption. Statewide inventories are given by EIA (2014) and CARB (2015) curves. Regional inventories include Hestia results and natural gas from power plants (CEC, 2014) in Los Angeles and Orange counties with monthly data (except Calabasas and Valencia). The vertical axes have been adjusted to allow easy comparison. This analysis is consistent with the increase in natural gas usage during the last few years.

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**Figure A1.** Miller-Tans plots for each month in 2011. Values of the slopes for three-month seasonal averages are plotted in Fig. 6a.

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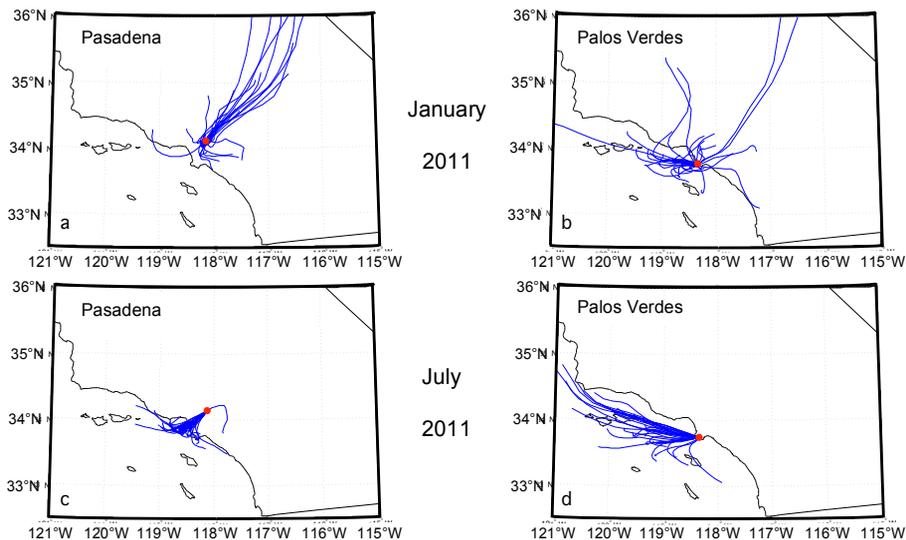
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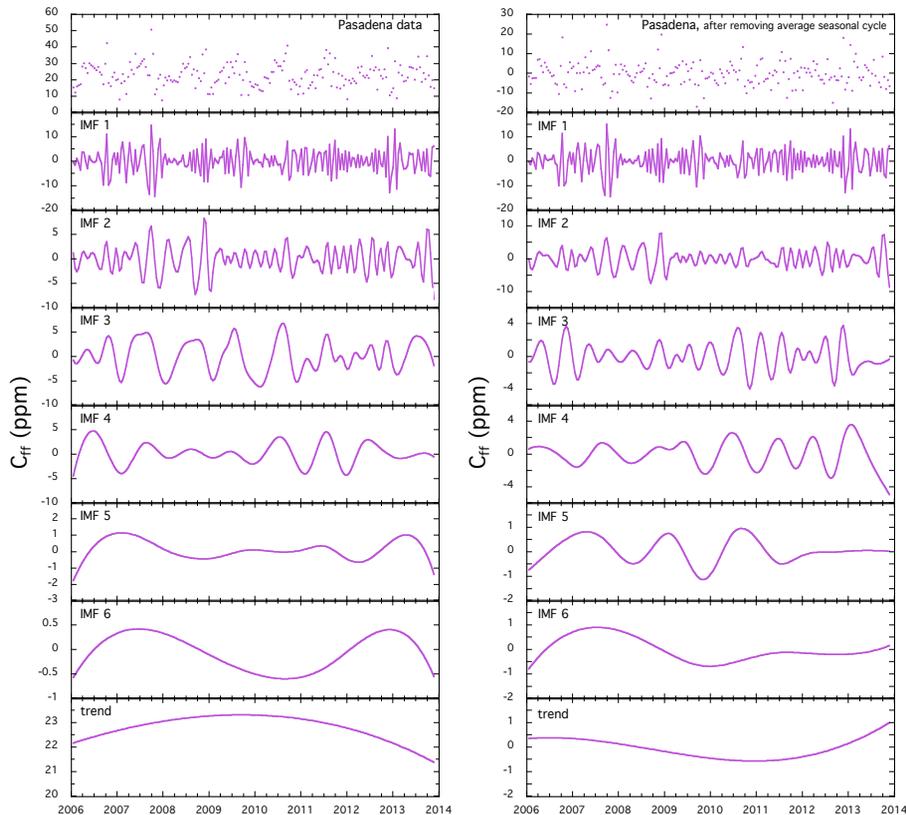
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**Figure A2.** Twelve-hour back trajectories for all days in January and July 2011, for the Pasadena and Palos Verdes sites. This shows more detail for the effect of transport on the air masses sampled during summer and winter at the Palos Verdes site than Fig. 7.

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**Figure A3.** Time series of all of the results from the ensemble empirical mode decomposition (EEMD) analysis of the Pasadena  $C_{ff}$ . The left set of panels shows the results for the raw data, whereas the right column shows those for the data after subtraction of the average seasonal cycle. The long-term trend reflecting the economic downturn of the Great Recession is reflected clearly in IMF 6 and the trend of the data after the pronounced seasonality is removed (right-hand column), although there is some evidence of it in IMF 6 of the raw  $C_{ff}$  data.